Argonne National Laboratory

UNCLASSIFIED QUARTERLY PROGRESS REPORT
ON PHYSICAL SCIENCES

August, September, October 1961

ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois

UNCLASSIFIED QUARTERLY PROGRESS REPORT
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Preceding Reports:

ANL-6394   August 1961
ANL-6349   May 1961
ANL-6298   February 1961

TABLE OF CONTENTS

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November 1961

This report presents briefly the unclassified accomplishments in the basic physical science program (5000 Program) of the Laboratory. It is a collection of abstracts of papers on work in the physical sciences published or accepted for publication since August 1961.

Research activities which have not yet reached the state of accepted journal publications are not covered. Neither is classified work covered; this will be reported in a series of supplementary reports which will be issued at less frequent intervals.

PHYSICS DIVISION

Theoretical Physics

ACCURACY OF THE SUPERCONDUCTIVITY APPROXIMATION FOR
PAIRING FORCES IN NUCLEI
A. K. Kerman, R. D. Lawson and M. H. MacFarlane
Phys. Rev. 124 162-167 (October 1, 1961)

The accuracy of the superconductivity approximation for pairing
forces in nuclei is studied. To this end we have exactly diagonalized the
pairing force for certain nuclei and compared the results with those ob-
tained from the approximate calculation of Kisslinger and Sorensen. When
the energy is computed by use of the approximate wave function, which is
not an eigenfunction of the number operator, it is found that the excitation
energies of the low-lying states with seniority one and two are correct to
within 200 kev, whereas the ground-state energies are usually not given to
better than 500 kev. The wave function obtained by projecting out and
normalizing that part of the variational trial function that corresponds to
the correct number of particles is found to agree closely with the exact
energy eigenfunction. Overlap integrals greater than 98% are found in all
cases considered. The expectation values of the pairing Hamiltonian with
respect to these projected wave functions are therefore in excellent agree-
ment with the exact energy eigenvalues. The variational aspects of the
superconductivity approximation are also discussed briefly.

MAGNETIC MOMENTS OF MIRROR NUCLEI
Dieter Kurath
Phys. Rev. 124 552-554 (October 15, 1961)

The sum of the magnetic dipole moments for pairs of mirror nuclei,
as calculated with intermediate coupling functions in the 1p shell, is found
to be very insensitive to the degree of spin-orbit coupling. This property
can be understood as being due to the rotational nature of the wave functions,
and can also be interpreted in terms of wave functions from the Nilsson
model. Magnetic dipole moments are predicted for those nuclei which have
not been measured.

PROPERTIES OF NUCLEI WITH NEUTRONS AND PROTONS IN THE $1f_{7/2}$ SHELL

R. D. Lawson
Phys. Rev., in press

Elliott's generating procedure is used to derive wave functions for nuclei in which the $1f_{7/2}$ level is filling with neutrons and protons. It is assumed that the low-lying states of these nuclei have well defined isotopic spin and that the configuration is pure. This representation, which mixes states of different seniority, is used to calculate beta-decay transition probabilities, magnetic-dipole and electric-quadrupole properties of nuclei. The results are in much better agreement with experiment than are those obtained by use of a seniority classification for the nuclear states. In particular, there is a K selection rule which explains the anomalously long half-life for the $Ca^{47} \rightarrow Sc^{47}$ and $Ca^{45} \rightarrow Sc^{45}$ beta decays. The theoretical electric-quadrupole matrix elements are too small by a factor of 3 to 5 and the theoretical M1 lifetimes too short by approximately a factor of 10. For three particles in the $1f_{7/2}$ level (Sc^{43}) or three neutrons in the $1g_{9/2}$ state, the eigenfunctions given by the generator formalism are found to be almost identical with those derived from a conventional shell-model calculation.

DISPERSION OF GYROMAGNETIC RATIOS IN COMPLEX SPECTRA

Norbert Rosenzweig and C. E. Porter*
Phys. Rev. <u>123</u> 853-855 (August 1, 1961)

SYMMETRY THEORY OF THE AHARONOV-BOHM EFFECT: QUANTUM MECHANICS IN A MULTIPLY CONNECTED REGION

L. J. Tassie and Murray Peshkin
Ann. Phys., in press

The conventional Schrödinger treatment of the Aharonov-Bohm effect of inaccessible magnetic fields postulates the use of single-valued wave functions, even in a multiply connected region. The principles of symmetry theory are shown to justify this postulate in a cylindrically symmetric system and to lead directly to the Aharonov-Bohm effect. The necessity for the canonical angular momentum to be an integer or half-integer is the crux of the argument.

*Brookhaven National Laboratory

NEUTRON SCATTERING BY THE COMPLEX HARMONIC OSCILLATOR
POTENTIAL
  J. Sokoloff
    Phys. Rev., in press

A complex harmonic oscillator potential is used in an attempt to fit
low-energy neutron scattering and absorption data. Differential scattering
cross sections are compared with experiment for 1-Mev incident neutrons
and a wide spectrum of target neuclei. Total cross sections are compared
with experiment at 350 kev, 1 Mev, and 1440 kev for an extensive mass
spectrum. For the parameters used in the calculations, the complex har-
monic oscillator seems to share the shortcomings of the "best-fit" complex
square well.

INTERFERENCE EFFECTS OF RADIOFREQUENCY FIELDS IN RESONANCE
RADIATION
  M. N. Hack
    Nuovo Cimento, in press

The theory developed previously to investigate the splitting of
Zeeman lines by rf magnetic fields leads directly to the Bressel-Bitter
resonance obtained in experiments on "optical pumping." An extension of
the theory accounts for the effects of the modulation of light recently ob-
served in these experiments.

Experimental Physics

HIGH-FREQUENCY PLASMOIDS
  A. J. Hatch
    Proc. 5th Intern. Conf. on Ionization Phenomena in Gases, Munich,
    Aug. 28-Sept. 1, 1961, in press

Rf potentials have been measured with probes as functions of axial
position in high-frequency plasmoids. The plasmoids are established in
dry air at pressures in the range from 0.2 to 1 micron Hg between plane
parallel aluminum electrodes, 23 cm in diameter and 15 cm apart, at a
frequency of 15 Mc/sec. The most significant result is that the 15-Mc/sec
potential in the plasmoid is observed to be 180° out of phase with the applied
potential. This and other results support the hypothesis that longitudinal
oscillations of electrons in the plasma play a dominant role in the plasmoid
mechanism and lead to an explanation of the dark sheath.

ENERGY DEPENDENCE OF THE $B^{10}(d,p)B^{11}$ ANGULAR DISTRIBUTION

Benjamin Zeidman, J. L. Yntema and G. R. Satchler*

The angular distributions of the $B^{10}(d,p)B^{11}$ ground-state transition have been measured at incident energies of 12.5, 15.5, 18.5, and 21.5 Mev. The angular distributions at the higher energies cannot be fitted with Butler's theory for (d,p) reactions. Excellent fits have been obtained with distorted-wave theory. There are indications that it is necessary to use an energy-dependent deuteron potential. Non-stripping processes appear to make very little contribution to this reaction.

NUCLEAR ENERGY LEVELS OF Na^{24} IN THE REGION 630 TO 860 KEV

C. T. Hibdon

An investigation of the nuclear level structure of Na^{24} for neutron energies in the range from 630 to 860 kev revealed the presence of 73 levels distributed throughout the range. Each of the two previously known large peaks was found to be composed of a set of levels. These levels are attributable to values of J up to 7 for the first peak and up to J = 6 for the other one. No s-wave levels were identifiable in this region, and it is doubtful that any p-wave levels are present. A set of parameters was determined for the levels by a best fit to the data. These levels were then grouped with those up to 630 kev to obtain a combined total of 230 levels up to 860 kev. The parameters of these levels show the following distributions: (a) an approximate exponential distribution for the level spacings, (b) an approximate exponential or Porter-Thomas distribution for the neutron widths with a tendency to favor the latter, and (c) a distribution of the angular momenta which agrees with the theoretical distribution given by Bloch. Reasonable values of the strength functions were obtained for s- and p-wave levels (0.035 and 0.27, respectively), but the values for $\ell \geq 2$ appear to be much too large. A plot of the number of levels having energies $\leq E_n$ as a function of E_n shows fluctuations about a linear trend, with no bending away from this trend at high energies to indicate a general missing of levels.

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*Oak Ridge National Laboratory

ANGULAR CORRELATIONS IN INELASTIC SCATTERING OF PROTONS FROM Mg^{24}

T. H. Braid, J. L. Yntema and Benjamin Zeidman

Angular correlation measurements in the reaction $Mg^{24}(p,p'\gamma)Mg^{24}$ at $E_p = 10.7$ Mev show strong correlations at $\theta_p = 30°$, $60°$ and $90°$. Comparison with the predictions of the plane-wave and adiabatic-scattering models indicates a predominant direct-interaction contribution to the scattering at these angles. At $\theta_p = 120°$ and $150°$ there is no agreement with these predictions.

THE (d,He^3) REACTION NEAR $Z = 28$

J. L. Yntema, T. H. Braid, Benjamin Zeidman and H. W. Broek

The (d,He^3) reaction produced by 21.6-Mev deuterons on several targets in the Fe-Ni region gives information on the proton configurations of the ground states of the target nuclei, and in certain cases on the neutron configurations and excited states. The observed pickup of p-protons and Ni isotopes indicates excitation of the $f_{7/2}$ proton core. The experiments show that the neutron configurations of the ground states of Co^{59} and Fe^{58} are probably dissimilar. Admixtures of $\ell = 1$ components are observed in the transitions to the first excited states of Ni^{64} and Ni^{62}. This indicates either excitation of the $f_{7/2}$ proton core or an admixture of neutron configurations coupled to spin 2 in the Cu isotopes.

THE (α,t) REACTION ON NUCLEI

J. L. Yntema

The (α,t) reaction has been investigated for a number of nuclei. All transitions observed tend to peak in the forward direction. However, significant differences between known $\ell = 1$ and $\ell = 3$ transitions were observed. The (α,t) reaction proceeds very strongly where the transition involves high ℓ values. The positions of peaks observed in the $Co^{59}(\alpha,t)Ni^{60}$ reaction shows a very strong correlation with states preferentially excited in the inelastic scattering of α particles by Ni^{60}. A similar result has been observed for $Cu^{63}(\alpha,t)Zn^{64}$.

ELASTIC AND INELASTIC SCATTERING OF 43-MEV α PARTICLES IN THE Ni REGION

H. W. Broek, T. H. Braid, J. L. Yntema and Benjamin Zeidman

Proc. Rutherford Jubilee Conf., Manchester, Sept. 4-8, 1961, in press

Angular distributions for inelastic scattering to the 1.33-Mev (2^+), 4.1-Mev (3^-), and 5.1 Mev (4^+) states of Ni^{60} are fitted to the distorted-wave Born-approximation calculation of Rost and Austern, and distortion parameters are derived. The 2.5-Mev (4^+) state shows an angular distribution which differs widely from the $\ell = 4$ curve. Differences in elastic scattering from the various nuclei are noted and ascribed to possible variation of surface thickness. Excitation of the prominent 2^+ groups varies by a factor of 5 with no apparent regularity, while excitation of the 3^- groups tends to increase with increasing A.

STUDIES OF (d,t) REACTIONS ON THE ISOTOPES OF NICKEL

M. H. Macfarlane, B. J. Raz, J. L. Yntema and Benjamin Zeidman

Proc. Rutherford Jubilee Conf., Manchester, Sept. 4-8, 1961, in press

Angular distributions and absolute cross sections have been measured for triton groups resulting from (d,t) reactions on Ni^{58}, Ni^{60}, and Ni^{61} at a deuteron energy of 21.6 Mev. The Butler theory is used, as previously described, to identify ℓ values and extract reduced widths. The reduced widths are then interpreted in terms of the structure of the nuclear states involved. The results of the analysis are that the occupation number for $1f_{5/2}$ neutrons is between 0.3 and 0.6 for Ni^{58}, 0.8 and 1.6 for Ni^{60}, and ≈ 1 for Ni^{61}. The results for Ni^{58} and Ni^{60} are in good agreement with predictions of the pairing model.

NEUTRON EMISSION FROM COMPOUND NUCLEAR SYSTEMS OF HIGH ANGULAR MOMENTUM

H. W. Broek

Phys. Rev. 124 233-245 (October 1, 1961)

Abstract in ANL-6394, p. 9

ISOTOPIC IDENTIFICATION OF NEUTRON RESONANCES OF Cd, Sb, Os
AND Pt FROM CAPTURE GAMMA-RAY SPECTRA
H. E. Jackson and L. M. Bollinger

The Argonne fast chopper has been used in conjunction with a
3-variable magnetic-tape recording system to demonstrate the high effi-
ciency and sensitivity of isotopic identification of neutron resonances by
observation of capture γ rays. A discussion of features of capture spectra
which can be useful in isotopic assignment is presented as well as results
for resonances in natural Cd, Sb, Os, and Pt. The experimental study re-
ported emphasizes primarily the observation of coincident γ rays as a
means of measuring binding energies and characteristic low-energy lines
of nuclides giving rise to resonant capture. The isotopic assignments in-
clude 23 resonances in Cd between 18 ev and 235 ev, 6 of which were pre-
viously unassigned; 20 resonances in Sb between 5 and 200 ev, 23 resonances
in Os in the region between 6 ev and 61 ev; and two previously unresolved
resonances at 67 ev assigned to Pt^{195}. The data lead to observed level
spacings of 33 ev in Cd^{111}, 80 ev in Cd^{112}, 27 ev in Cd^{113}, 27 ev in Sb^{121}, 12 ev
in Sb^{123}, and 5.1 ev in Os^{189}. Strength functions for Cd^{111} and Cd^{113} based on the
identifications are in agreement with previous reported values. For Os^{189},
from transmission data previously unreported, the value $\Gamma_n^0/D = (2.0^{+2.2}_{-0.7})$
x 10^{-4} is obtained.

CAPTURE-GAMMA-RAY SPECTRUM OF $Cd^{113}(n,\gamma)Cd^{114}$ AND THE
ASSOCIATED ENERGY LEVELS IN Cd^{114}
R. K. Smither

The capture gamma-ray spectrum of $Cd^{113}(n,\gamma)Cd^{114}$ was investigated
with the Argonne 7.7-meter bent-crystal spectrometer. The observed
spectrum consisted of 119 gamma rays with energies below 2 Mev. These
precision energy measurements were combined with a series of coinci-
dence experiments to modify and extend the level scheme of Cd^{114}. The
errors in the energy values of five previously observed levels are reduced
by a factor of 10. Their new values are found to be 557.8 $\pm$ 0.1, 1208.4 $\pm$ 0.2,
1282.2 $\pm$ 0.2, 1304.9 $\pm$ 0.3, and 1362.9 $\pm$ 0.3 kev. Five new levels are estab-
lished at 1133.1 $\pm$ 0.2, 1730.3 $\pm$ 0.2, 1839.9 $\pm$ 0.3, 1958.3 $\pm$ 0.3, and
2202.4 $\pm$ 0.3 Mev. Eight more levels are suggested at 1607.8 $\pm$ 0.3,
2048.3 $\pm$ 0.4, 2225.1 $\pm$ 0.4, 2392.5 $\pm$ 0.4, 2573.8 $\pm$ 0.5, 2868.0 $\pm$ 0.7, 3216.8 $\pm$ 0.8,
and 3484.7 $\pm$ 0.8 kev. The limits placed on the spins and parities of these
levels by the observed capture gamma rays are discussed.

SPECTROSCOPY OF GAMMA RADIATION FROM Nd^{144}, Sr^{88} AND Pb^{207}
   J. E. Monahan, Sol Raboy and C. C. Trail
      Phys. Rev. 123 1373-1381 (August 15, 1961)

   Abstract in ANL-6394, p. 10

ENERGY LEVELS IN $_{69}Tm^{172}$ FROM THE DECAY OF $_{68}Er^{172}$
   R. G. Helmer and S. B. Burson
      Phys. Rev. 123 992-996 (August 1, 1961)

   Abstract in ANL-6298, p. 19

DECAY OF Tm^{172}
   R. G. Helmer and S. B. Burson
      Phys. Rev. 123 978-991 (August 1, 1961)

   Abstract in ANL-6298, p. 19

PRINCIPLES OF CYCLIC PARTICLE ACCELERATORS
   J. J. Livingood
      New York: D. Van Nostrand Co., 1961

   Abstract in ANL-6349, p. 19

SPIN ASSIGNMENT OF RESONANCES FROM CAPTURE GAMMA-RAY
SPECTRA
   L. M. Bollinger and R. E. Coté
      Proc. Symp. on Neutron Time-of-Flight Methods, Saclay, July 24-26,
      1961, in press

      The previous proposal for the determination of the spins of neutron
resonances by the observation of sum-coincidence γ-ray spectra is explored
further. The basic idea of the proposed method is that, for a large class of
nuclides, the intensity of γ-ray cascades that proceed in exactly two steps
from the initial compound state to some particular low-lying state will de-
pend on the spin of the initial state. By use of the Argonne 3-parameter
magnetic-tape recording analyzer, the sum-coincidence spectra of Mo^{95}, Cd^{111},
Cd^{113}, Hg^{199}, and Hg^{201} are investigated in detail. The measurements are
shown to be technically feasible for all these nuclides. Moreover, for Mo^{95},
Cd^{111}, and Cd^{113}, all of which form states with positive parity, the spectra
appear to satisfy the statistical requirements that are necessary to make the
method useful. However, for Hg^{199} and Hg^{201}, so few modes give significant
contributions of 2-step cascades to the ground state that statistical fluctua-
tions may comprise the reliability of the 2-step-cascade method. Nevertheless,
the γ-ray spectra give spin assignments that are consistent with those obtained
in other ways.

LESSONS LEARNED FROM A CHOPPER ACCIDENT

G. E. Thomas, R. E. Coté and L. M. Bollinger

Recently an effort was made to increase the speed of rotation of Rotor II of the Argonne fast chopper to 20,000 rpm. At some speed between 19,000 and 20,000 rpm, the rotor suffered a serious non-elastic deformation. It is believed that the deformation of the Monel and uranium sections in which the slits were cut might have been less if the bottom plate had been strong enough. The main purpose of this paper is to describe the unusual technique used to repair the Monel and uranium parts. Briefly, the technique consisted of compressing the parts by means of the thermal contraction of a closely fitting steel ring. Measurements on the rotor after a series of these operations indicated that most of the dimensions had been restored to their original values, and subsequent use of the restored rotor indicates that the operation was completely successful.

ISOTOPIC IDENTIFICATION OF RESONANCES FROM CAPTURE GAMMA-RAY SPECTRA

H. E. Jackson and L. M. Bollinger

The use of capture γ rays for the isotopic identification of neutron resonances has been studied systematically. The properties most generally applicable for such identification are found to be the characteristic low-energy γ rays and the neutron binding energy. Both properties are shown to be most sensitively observed by the detection of the spectra associated with coincident counts in two large NaI(Tl) scintillators. Spectra of this kind are recorded for the resonances of cadmium, antimony, osmium, and platinum. On the basis of these measurements, it is concluded that a) the gamma-ray method is much faster and more sensitive than are transmission measurements of separated isotopes for most of the even-odd target nuclides, b) it is a competitive technique for other nuclides if the spacings between low-lying levels of the compound nucleus are sufficiently different, and c) it is inferior to the transmission method when the low-energy levels of the compound nucleus are too close together and when, simultaneously, the binding energies of the nuclides are not sufficiently different.

ANALYSIS OF RESONANT-CAPTURE γ-RAY SPECTRA

R. T. Carpenter, J. P. Marion and L. M. Bollinger
Proc. Symp. on Neutron Time-of-Flight Methods, Saclay, July 24-26, 1961, in press

Techniques for quantitative analysis of the γ-ray spectra from capture of neutrons in resonances are described. In the most general case, the γ-ray spectra from all observed resonances in a given nuclide are treated simultaneously. A least-squares fit is performed to obtain the energies of a given number of γ-ray lines (which are the same for all resonances in a given nuclide), the intensities of the transitions (which vary from resonance to resonance), and the statistical errors associated with these quantities. The simultaneous treatment minimizes the errors in the energies. The treatment can be reliably applied to spectral regions in which the lines are partially resolved in any of the resonances. The reference line shapes, which are allowed to depend on the γ-ray energy, are calculated by interpolation between line shapes given to the computer in tabular form for single transitions at various energies. Any physical knowledge of the intensities and energies of any of the lines in the spectra may be fed into the problem as linear constraints. For example, one often knows the difference in energy between at least some of the lines. A χ^2 test of the goodness of fit gives a check on the assumptions used in the calculation. Various special cases of the most general calculation are permitted. The calculations are performed on the IBM-704 digital computer.

GLASS SCINTILLATORS FOR NEUTRON DETECTION

L. M. Bollinger and G. E. Thomas
Proc. Symp. on Neutron Time-of-Flight Methods, Saclay, July 24-26, 1961, in press

The characteristics of lithium-containing glass scintillators made by Ginther and boron-containing scintillators made by Ginther and Bishay are discussed with reference to the detection of neutrons. Experimental results on the dependence of pulse height on composition are presented. Measured curves for the dependence of scintillation intensity on time are given. The measured time-dependence and height of the scintillation pulses and the calculated efficiency of neutron capture and the capture time are considered in relation to the various possible applications of the scintillators to neutron detection.

A γ-RAY SPECTROMETER FOR STUDIES OF RESONANT-CAPTURE
γ-RAY SPECTRA
L. M. Bollinger, R. T. Carpenter, R. E. Coté and H. E. Jackson
Proc. Symp. on Neutron Time-of-Flight Methods, Saclay, July 24-26,
1961, in press

A description is given of the characteristics of a large NaI(Tl) scintillator that is particularly well suited to the study of γ rays from capture in neutron resonances. The crystal is 8 inches in diameter and 6 inches thick. It is coupled to a single 9545A EMI photomultiplier through a short Lucite light pipe. The resulting system gives a pulse-height resolution width of only 3.5% for 8-Mev γ rays. The line shape and its dependence on collimation and on the position and size of the source are discussed. An unexpected background peak is observed at an energy of about 6.9 Mev. The system is found to be extremely stable.

MEASUREMENT OF THE TIME DEPENDENCE OF SCINTILLATION
INTENSITY BY A DELAYED-COINCIDENCE METHOD
L. M. Bollinger and G. E. Thomas
Rev. Sci. Instr. 32 1044-1050 (September 1961)

A new method for the measurement of the time dependence of the light intensity from scintillators is described. An important aspect of the method is the ease with which it can be applied to all modes of excitation. The results of measurements on the time dependence of the scintillators for several modes of excitation are presented.

ION FORMATION ON HOT METAL SURFACES
Manfred Kaminsky
Physik. Blätter, in press

This paper was one in a series of invited lectures presented at the German Physical Society meeting, Bad Nauheim, April 20, 1961. It surveys the present state of research in surface ionization and gives a critical review of experimental methods, experimental data, and theoretical interpretations.

ON A MASS SPECTROMETER FOR DETECTING PULSED MOLECULAR BEAMS AND ITS USE FOR INVESTIGATING ADSORPTION AND DESORPTION PROCESSES ON METAL SURFACES

Manfred Kaminsky

The constructional features of a pulsed molecular beam apparatus are described; together with a mass spectrometer it will be used as detector in investigating the kinetic processes occurring in the interaction between gas and solids. The use of this facility for solving problems in other fields (as for instance, cathode sputtering, ion neutralization on surfaces, elastic reflection of atoms and ions from the metal surfaces) from this standpoint is not described in detail here.

THE MASS SPECTRUM OF ETHYL-LITHIUM VAPOR

Joseph Berkowitz, D. A. Bafus* and T. L. Brown*

Abstract in ANL-6394, p. 13

SOME EFFECTS OF TEMPERATURE AND VISCOSITY ON FLUORESCENCE AND ENERGY TRANSFER IN SOLUTIONS

Arye Weinreb

Abstract in ANL-6394, p. 13

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*University of Illinois

FLUORESCENCE ENERGY TRANSFER AND OXYGEN QUENCHING IN
SOLUTIONS OF DIPHENYLOXAZOLE IN CYCLOHEXANE
Arye Weinreb
J. Chem. Phys., in press

The fluorescent intensity of solutions of diphenyloxazole (PPO) in
cyclohexane is measured as a function of concentration for excitation by
γ rays. The fluorescence spectrum of the solutions when excited by uv is
recorded. These uv measurements, together with the results of β excita-
tion, were used to examine the quenching influence of oxygen on the solute
fluorescence and on the energy transfer from cyclohexane. The decay
times of the solutions are measured. The results are interpreted to show:
(1) a pronounced tendency of PPO to form molecular complexes in cyclo-
hexane, (2) a "static" character for the oxygen quenching of cyclohexane.
The dependence of transfer probability on solute concentration is
discussed.

PROGRESS IN PLASTIC SCINTILLATORS
L. J. Basile
Proc. Univ. New Mexico Conf. on Organic Scintillation Detectors,
Aug. 15-17, 1960. Ed. G. H. Daub, F. N. Hayes and Elizabeth
Sullivan. TID-7612, pp. 161-176.

Since the development of plastic scintillators in 1950 there has been
considerable information amassed on recipes for producing plastic scintil-
lators, luminescent properties of solutes in a plastic medium, effects of
the plastic medium on the mechanism of energy transfer in the scintillation
process, and the use of plastic scintillators to detect ionizing radiation.
Recent advances in each of these areas are described.

THE DESIGN OF BUNCHING MAGNETS
R. E. Holland
Nuclear Instr. & Methods 12 103-110 (June 1961)

Abstract in ANL-6349, p. 21

PARTICLE ACCELERATOR DIVISION

REMOTE BINARY CONTROL OF MULTIPLE MOTORS
Walter Leonchick and J. F. Mech
ISA Journal 8(8) 93-97 (August 1961)

Two problems in the remote actuation of one in many widely-separated electric motors were solved. The first problem called for the positioning of any one of 56 tuning balls of the linac cavity of the zero gradient synchrotron. The problem here was to select and control from a central location any one in 56 motors. The method devised employs binary control without relays, featuring binary-to-decimal reconversion for direct readout. The second problem involved remote selection of any 1 in 100 electric motors for both forward or reverse rotation; this was done using only 20 standard relays.

HIGH ENERGY PHYSICS DIVISION

USE OF SPARK CHAMBERS IN MAGNETIC FIELDS
G. R. Burleson, Arthur Roberts and T. A. Romanowski
Rev. Sci. Instr. 32 (1069-1070 (September 1961)   Letter

Abstract in ANL-6394, p. 16

ADDENDUM TO THE PAPER "DEVELOPMENT OF THE SPARK CHAM-
BER: A REVIEW"
Arthur Roberts
Rev. Sci. Instr. 32 996 (August 1961)   Letter

The author's attention has been called to a paper by G. Charpak,
published in 1957. In these experiments Charpak was successful in obtain-
ing photographs in which a pulsed spark discharge followed the track of
an alpha particle at angles up to about 45° to the electric field. Since these
experiments antedate those of Fukui and Miyamoto, it seems that they were
in fact the first successful attempts to produce a spark chamber operating
in the mode in which the tracks are delineated rather than sampled. The
chamber, using 10% propane in argon, was not successful with electrons,
however, and Fukui and Miyamoto still appear to be the first to achieve
successful delineation of minimum ionizing tracks.

RECENT WORK ON A CERENKOV IMAGE DETECTOR
G. R. Burleson, W. J. Kernan, Arthur Roberts and T. A. Romanowski
Proc. Nuclear Electronics Conf., Belgrade, May 15-20, 1961,
in press

Abstract in ANL-6394, p. 19

A SPARK CHAMBER SPECTROMETER
G. R. Burleson, Arthur Roberts and T. A. Romanowski
Proc. Nuclear Electronics Conf., Belgrade, May 15-20, 1961,
in press

Abstract in ANL-6394, p. 17

LODESTAR: A NEW COUNTER DATA HANDLING FACILITY

Arthur Roberts and Curtis Rockwood*

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*Electronics Division

APPLIED MATHEMATICS DIVISION

PARTIAL WAVE THEORY OF POSITRON-HYDROGEN ATOM COLLISION
Kenneth Smith
Proc. Phys. Soc., in press

This paper is concerned with the derivation of the radial equations for positron-hydrogen atom collisions from the continuous state Hartree-Fock equations. The angular coefficients are expressed in terms of vector addition coefficients and Racah coefficients for any angular momentum state of the atomic systems: positronium and atomic hydrogen.

The boundary conditions are written in terms of the S-matrix and expressions are obtained for total and differential cross sections.

THE SOLUTION OF MANY REGION REACTOR KINETICS PROBLEMS ON AN ANALOG COMPUTER
C. N. Kelber,* L. C. Just, and N. F. Morehouse, Jr.
Nuclear Sci. and Eng., in press

The time dependent reactor kinetics equations are put in a form to permit their solution on an analog computer. An example of a high flux reactor computation is then given.

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*Reactor Engineering Division

CHEMISTRY DIVISION

Nuclear Chemistry

EIGENVALUE PROBLEMS IN MATRIX MECHANICS

R. R. Chasman
J. Math Phys. 2 733-735 (September-October 1961)

The methods of the Heisenberg matrix mechanics are employed to treat the potential Q^4 in detail. Bound state energy levels and elements of the Q matrix are obtained. A semiquantitative treatment is given for the potential Q^6.

NEW YTTRIUM ISOMERS OF MASSES 86 AND 90

Larry Haskin and Robert Vandenbosch
Phys. Rev. 123 184-185 (July 1, 1961)

Abstract in ANL-6394, p. 24

NEUTRON CAPTURE CROSS-SECTION OF 36-HR Rh^{105}

J. G. Cuninghame, L. E. Glendenin and A. L. Harkness*
J. Inorg. and Nuclear Chem., in press   Letter

When mass spectrometric thermal fission yields of Pu^{239} were measured, the yield of Pd^{105} was observed to be low relative to those of Pd^{107}, Pd^{108}, and Pd^{110}, and the yield of Pd^{106} was correspondingly high, indicating a large neutron capture cross section for 36-hr Rh^{105}. For an estimated maximum neutron flux of 3×10^{14} $n/cm^2/sec$ the (n,γ) cross section for Rh^{105} is calculated to be $\geq 1.8 \times 10^4$ barns.

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*Special Materials Division

NEUTRON DIFFRACTION STUDIES OF La_2C_3, Ce_2C_3, Pr_2C_3 AND Tb_2C_3

Masao Atoji and D. E. Williams

J. Chem. Phys., in press

The neutron diffraction data of the rare earth sesquicarbides with the Pu_2C_3 type structure ($D5_c$-type), La_2C_3, Ce_2C_3, Pr_2C_3, and Tb_2C_3, were analyzed using the complete-matrix least-squares method. These sesquicarbides contain the C_2 groups with the C-C distances, 1.236 ± 0.009 A, 1.276 ± 0.005 A, 1.239 ± 0.003 A, and 1.240 ± 0.005 A, respectively, for La_2C_3, Ce_2C_3, Pr_2C_3, and Tb_2C_3. These values are, except for Ce_2C_3, significantly longer than the C-C distance in CaC_2, 1.191 A, but are shorter than the average C-C distance in the rare earth dicarbides, 1.278 A.

The paramagnetic scattering analyses show that all metal atoms in these sesquicarbides are in their trivalent Hund ground state, except Ce in Ce_2C_3, whose possible valency is about 3.4. The screening constants for the 4f hydrogenic radial wave functions of Pr and Tb as determined from the diffuse scattering analyses of their sesquicarbides are 40 and 43, respectively. The valency and metallic bonds in these sesquicarbides are briefly discussed.

SPECTROSCOPIC DETERMINATION OF THE NUCLEAR SPIN OF Lu[176]
AND THE NUCLEAR MAGNETIC AND QUADRUPOLE MOMENTS OF
Lu[175] AND Lu[176]

F. S. Tomkins, S. Gerstenkorn,* J. Blaise* and J. Bauche*
Atomic Spectroscopy Symp. Sponsored by National Research Council
Committee on Line Spectra and Argonne National Laboratory,
June 7, 8, and 9, 1961 at Argonne National Laboratory. Abstract of
Papers, pp. 19-20

The hyperfine structure of several lines of Lu I and Lu II, emitted
by a sample of lutetium containing 74.5% Lu[176] and 25.5% Lu[175], have been
recorded using a Fabry-Perot spectrometer. From the ratio of the inten-
sities of the hyperfine structure components, it is deduced that $I^{176} = 7$,
confirming the result of Kaliteevskii and Chaika. The ratios of the moments
of the two isotopes are:

$$\mu^{175}/\mu^{176} = 0.7109 \pm 0.0050 \text{ and } Q^{175}/Q^{176} = 0.71 \pm 0.01$$

The value proposed by Steudel, $\mu = +2.0 \pm 0.2 \; \mu_N$ is confirmed, but the
value of the quadrupole moment must be revised:

$$Q^{175} = (4.0 \pm 0.5) \cdot 1/(1-R) \cdot 10^{-24} = (5.6 \pm 0.6) \cdot 1/(1-R') \cdot 10^{-24} \text{ cm}^2$$

where R and R' are the Sternheimer corrections for the 6p and 5d electrons,
respectively. The results are compared with the predictions of the model
of Mottelson and Nilsson, the ratios of the quadrupole moments indicating
that the deformations of the two nuclei are identical.

NEW HAFNIUM ISOTOPE, Hf[182]
James Wing, B. A. Swartz and J. R. Huizenga

Abstract in ANL-6394, p. 25

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*C.N.R.S., Bellvue (S&O) France

FISSION FRAGMENT ANISOTROPY AND PAIRING EFFECTS ON NUCLEAR STRUCTURE

Robert Vandenbosch, H. Warhanek and J. R. Huizenga
Phys. Rev., in press

The dependence of the angular distributions on projectile energy for helium-ion-induced fission of Th^{232}, U^{233}, and U^{238} has been investigated using solid-state fission fragment detectors. The measurements cover the range of projectile energies between 20 and 43 Mev in steps of 1 Mev or less. The energy dependence of the anisotropy has a structure which is in qualitative agreement with theoretical predictions. The most fissionable target studied, U^{233}, shows the least structure while the least fissionable target, Th^{232}, shows the most structure, demonstrating that large anisotropies result from fission occurring at low excitation energy following neutron emission from the compound nucleus. The energy dependence of K_0, the standard deviation of the distribution in the angular-momentum projection on the nuclear symmetry axis at the saddlepoint, has been deduced from this and other data on angular distributions from neutron induced fission. At high excitation energies K_0 is consistent with predictions of statistical theory with a moment of inertia close to that of a rigid rotator. At excitation energies in excess of the fission threshold of 10 Mev or less, K_0 is much reduced due to nuclear pairing effects.

FAST-NEUTRON-INDUCED FISSION CROSS SECTIONS OF Pu^{241} AND Am^{243}

D. K. Butler* and R. K. Sjoblom
Phys. Rev., in press

The cross sections for neutron-induced fission of Pu^{241} for neutrons of energies from 0.02 to 1.80 Mev and of Am^{243} for neutrons of energies from 0.3 to 1.7 Mev have been determined. The determination was made by measuring the ratio of each cross section to that of U^{235} using a back-to-back gas scintillation counter.

PROMPT FISSION NEUTRON SPECTRUM OF Pu^{241}

A. B. Smith,* R. K. Sjoblom and J. H. Roberts**
Phys. Rev. 123 2140-2142 (September 15, 1961)

Abstract in ANL-6394, p. 25

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*Reactor Engineering Division

**Northwestern University

Effects of Radiation

MOLECULAR DETACHMENT PROCESSES IN THE VACUUM UV
PHOTOLYSIS OF GASEOUS HYDROCARBONS: I. ETHYLENE. II. BUTANE
M. C. Sauer, Jr. and L. M. Dorfman
J. Chem. Phys. 35 497-502 (August 1961)

Abstract in ANL-6394, p. 28

THE RADIOLYSIS OF ETHYLENE: DETAILS OF THE FORMATION OF
DECOMPOSITION PRODUCTS
M. C. Sauer, Jr. and L. M. Dorfman
J. Phys. Chem., in press

The radiolysis of ethylene has been studied with emphasis on the
decomposition products. The G-values of hydrogen and acetylene are
independent of ethylene pressure from 150 to 1000 mm, being 1.2 and
2.4 molecules per 100 ev respectively, but increase significantly below
this pressure. Isotopic and scavenger data are presented for the system
C_2H_4 - C_2D_4.

The data indicate that hydrogen is formed by molecular detachment
from an excited ethylene molecule or molecule ion, and that acetylene is
formed by molecular detachment from an excited ethylene molecule and/or
an ion molecule reaction involving $C_2H_3^+$.

The data do not allow us to specify the relative importance of ionic
and excited states in the formation of these products. However, the pres-
sure independence of the yields above 150 mm indicates that, in this region,
a competition between unimolecular dissociation and ion-molecule reactions
of the parent ion is highly unlikely

RADIATION CHEMISTRY: INTERMEDIATES IN LIQUIDS
L. M. Dorfman
Nucleonics 19 (10) 54-56 (October 1961)

This is a review article of work in the recently developed field of detection of intermediates in irradiated liquids.

Pulse radiolysis has become feasible within the past few years through the availability of electron accelerators which deliver a very intense, short pulse of high energy electrons. Observation of the decay of the transient species formed has provided information on the elementary processes.

At Argonne, for example, pulse radiolysis studies of aqueous solutions of benzene have yielded a transient uv absorption spectrum which has subsequently been assigned to the hydroxy cyclohexadienyl radical, HOC_6H_6. The absolute rate constant for the reaction of this radical with O_2 was measured as $k = 5.0 \times 10^8 \ \ell \ m^{-1} \ sec^{-1}$. A second spectrum obtained in oxygenated solution is attributed to the organic peroxy radical. Transient absorption spectra have also been observed in aqueous solutions of inorganic compounds. The diiodide ion, I_2^- has been observed in pulse radiolysis of dilute potassium iodide solutions in water, and the pulse radiolysis of potassium bromate yields a spectrum attributable to the bromate radical, BrO_3.

At Mellon Institute, the detection by electron spin resonance of molecular species and fragments containing unpaired electrons has been applied to the identification and measurement of steady-state concentrations of organic free radicals in irradiated liquids. Observations have been made in liquid ethane and in many other irradiated liquid hydrocarbons.

At the University of Notre Dame, a recent advance in the measurement of fast luminescence decay times is the use of the image converter tube in a repetitive time selection technique, permitting a time resolution shorter than 10^{-9} sec.

These methods are providing absolute rate constant values and more direct information on the elementary processes in irradiated liquids.

CHEMICAL YIELDS OF IONIZING RADIATIONS IN AQUEOUS SOLUTIONS: EFFECT OF ENERGY OF ALPHA PARTICLES
Sheffield Gordon and E. J. Hart
Radiation Research 15 440-451 (October 1961)

Abstract in ANL-6394, p. 26

RADIATION CHEMISTRY OF WATER WITH PULSED HIGH INTENSITY ELECTRON BEAMS

A. R. Anderson and E. J. Hart
J. Phys. Chem., in press

The radiation chemistry of water, aqueous ferrous sulfate, hydrogen peroxide and formic acid has been studied using pulsed electron beams from the Argonne linear electron accelerator. With a pulse length of ~1.4 μsec and an electron energy of ~15 Mev, the dose received by the solutions during a pulse is equivalent to ~2 x 10^{23} ev ml^{-1} sec^{-1}. All irradiations are monitored with the Fricke dosimeter, for which $G(Fe^{3+})$ is 11.4 $\pm$ 0.5 under these irradiation conditions. Preliminary studies with neutral water and with 0.8 $\underline{N}$ sulfuric acid solutions show that the initial yields of both hydrogen and hydrogen peroxide are about 80% higher in the acid solution but that, in each case, $G(H_2) = G(H_2O_2)$.

Yields of hydrogen have been measured at hydrogen peroxide concentrations up to 8 $\underline{M}$ in acid solution and up to 0.1 $\underline{M}$ in neutral solution. In each case, but at different concentrations of hydrogen peroxide, the scavenging curves eventually coincide with those determined using Co^{60} γ radiation at a dose rate of ~10^{17} ev ml^{-1} sec^{-1}. These data are consistent with the transient existence of two reducing species designated as the solvated electron, e_{aq}^-, predominant in neutral solutions, and the H atom, predominant in acid solution. Relative rate constants consistent with the observed hydrogen and hydrogen peroxide yields are:

$k_{H+H}/k_{H+H_2O_2}$ = 150; $k_{H+H}/k_{e_{aq}^-+H_2O_2}$ = 3; $k_{e_{aq}^-+H_2O_2}/k_{H+H_2O_2}$ = 50; $k_{e_{aq}^-+H_2O_2}/k_{OH+H_2}$ ~100; k_{H+H} = 2k_{OH+OH} = 0.5 k_{H+OH} = 0.5 k_{H+O_2} = 0.5k_{OH+HO_2} = 0.1$k_{e_{aq}^-+OH}$.

In contrast to the lower $G(Fe^{3+})$ obtained with ferrous sulfate, measurements with the formic acid dosimeter (0.01 $\underline{M}$ HCOOH, 0.001 $\underline{N}$ H_2SO_4, 0.001 $\underline{M}$ O_2) give the same yields as with Co^{60} γ radiation at much lower dose rates. The hydrogen peroxide yield for oxygen saturated neutral water is higher than that for Co^{60} γ radiation and can be explained on the basis of competing radical-radical reactions.

RADIATION CHEMISTRY: WATER

E. J. Hart

Studies of water ionization and excitation by radiation have yielded information on hydrogen and hydroxyl free radical yields and diffusion kinetics and have resulted in identification of species generated in irradiated water. As more is learned about reactions in irradiated water it becomes clear that all reactions are not explained by free-radical mechanisms. Among the several additional species postulated, the solvated electron or polaron is important. Irradiation of water by electrons, x-rays and gamma rays produces hydrogen and hydroxyl free radicals, whereas heavy particles produce large amounts of molecular hydrogen and hydrogen peroxide. Simultaneous irradiation of water by combinations of gamma rays and protons or electrons gives additive effects for simple nonchain reactions, but chain reactions behave differently. In the irradiation of inorganic aqueous solutions, the large reducing power of hydrogen and the large oxidizing power of the hydroxyl radical promote chemical changes in dissolved molecular or ionic species. The detailed free-radical mechanisms in the reactions of hydrogen and the hydroxyl radical with organic molecules are still largely speculative.

RADIATION CHEMISTRY

E. J. Hart and R. L. Platzman*

Abstract in ANL-6298, p. 36

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*Radiological Physics Division

RADIATION CHEMISTRY: INTERMEDIATES IN SOLIDS
M. S. Matheson
Nucleonics 19 (10) 57-60 (October 1961)

Many chemical intermediates ordinarily unstable in a liquid or a
gas can be preserved and measured in a solid if the solid prevents diffusion
and does not react with the trapped species. Methods employed include
electron-spin resonance (ESR), ultraviolet absorption spectroscopy, infra-
red absorption (for stable products only) fluorescence, phosphorescence
and thermoluminescence, x-ray diffraction (for matrix structure only),
differential thermal analysis and chemical methods. Intermediates have
been found in irradiated aliphatic hydrocarbons, alcohols and other organic
compounds of small molecular weight - especially by the use of ESR.
Since water is an important compound in radiation chemistry, trapped
intermediates in ice have been investigated thoroughly. The principal
radicals that explain the radiation chemistry of water have been found in
irradiated ice. A number of irradiated polymers have been examined for
induced electrical conductivity and electron-spin resonance and theories
have been advanced to explain some of the observations.

IRRADIATED LITHIUM FLUORIDE AS AN OPTICAL FILTER IN THE
FAR ULTRAVIOLET
J. L. Weeks, Sheffield Gordon and G. M. A. C. Meaburn
Nature 191 1186-1187 (September 16, 1961) Letter

Photochemical studies are being carried out using the 1849 A
radiation emitted by a low-pressure mercury resonance lamp. It has
been found necessary to eliminate the contribution of the intense 2537 A
line emitted by this source. The absorption spectrum of γ-irradiated
lithium fluoride shows a sharp band with a maximum near 2500 A due to
the formation of F-centers in the crystal. When this material was used
as the filter for mercury resonance radiation, the transmission of the
irradiated crystal remained high at 1849 A despite the complete absorp-
tion of the 2537 A line.

Inorganic and Physical Chemistry

INTERPRETATION OF THE INFRARED SPECTRA OF INORGANIC
MOLECULES
J. R. Ferraro
Proc. 5th Ann. Infrared Spectroscopy Inst., Canisius College,
Buffalo, April 14-18, 1961, in press

The infrared spectra of many inorganic salts have been determined
in the sodium chloride region and the cesium bromide region. From these
studies, several characteristic frequencies for polyatomic ions have been
found, and these have proved useful in qualitative inorganic analysis. How-
ever, the frequencies listed in various structure-spectra correlation charts
are for the ionic salts. There are many instances in which the polyatomic
anion (NO_3^-, SO_4^-, CO_3^-, IO_3^-) becomes coordinated to the cation, and this
causes a lowering of the symmetry of the anion. These are anions that
have double or triple degenerate vibrations, and the coordination is re-
flected in the infrared spectrum by additional absorptions because of the
removal of the degeneracy. Examples are the transition metal complexes
involving the above anions. Characteristic frequencies for polyatomic
inorganic anions are summarized for the most important anions.

CORRECTION OF THE POTASSIUM VAPOR PRESSURE EQUATIONS BY
USE OF THE SECOND VIRIAL COEFFICIENT
R. J. Thorn and G. H. Winslow
J. Phys. Chem. 65 1297-1302 (August 1961)

Abstract in ANL-6394, p. 30

ENERGY VALUE OF THE OCTAHEDRAL-TETRAHEDRAL COORDINA-
TION CHANGE
L. I. Katzin
J. Chem. Phys. 35 467-472 (August 1961)

Abstract in ANL-6394, p. 31

STRUCTURE OF THE BAND SPECTRUM OF CuCl MOLECULE. I. ADDI-
TIONAL KNOWLEDGE IN THE COARSE STRUCTURE
P. R. Rao and J. K. Brody
J. Chem. Phys. 35 776-787 (September 1961)

Abstract in ANL-6394, p. 31

EVIDENCE FOR Ga(I) AS AN INTERMEDIATE IN THE REACTION OF
GALLIUM METAL WITH AQUEOUS PERCHLORIC ACID

K. R. Schug and A. Sadowski*
J. Am. Chem. Soc. <u>83</u> 3538 (1961)   Letter

The observed stoichiometry of the reaction of gallium metal with
perchloric acid provides strong evidence for the participation of Ga(I) as
an intermediate species in the reaction, and the following mechanism is
proposed:

$$Ga(O) + H^+ = Ga(I) + \frac{1}{2} H_2(g)$$

$$4\ Ga(I) + 8H^+ + ClO_4^- = 4\ Ga^{3+} + Cl^- + 4\ H_2O \quad .$$
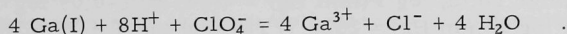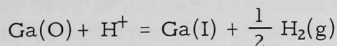
This mechanism involves highly selective behavior of Ga(O) and
Ga(I) with respect to oxidation, the metal reacting predominantly with H^+
rather than ClO_4^- or the intermediate chlorine compounds, and the Ga(I)
reacting predominantly with ClO_4^- instead of with H^+. On the basis of this
interpretation, gallium(I) joins the small group of ions capable of reducing
perchlorate ion in aqueous solution. The reluctance of Ga(I) to reduce H^+
provides an explanation for the slow rate of dissolution of the metal in the
halogen acids as compared with $HClO_4$ and for the observed reducing
properties of the solution after the metal is completely dissolved.

PRIMARY AND SECONDARY DEUTERIUM ISOTOPE EFFECTS ON RATE
OF MUTAROTATION OF FULLY DEUTERIATED GLUCOSE AND
MANNOSE

N. C. Li, Arlene Kaganove, H. L. Crespi and J. J. Katz
J. Am. Chem. Soc. <u>83</u> 3040-3043 (July 20, 1961)

Abstract in ANL-6394, p. 43

RHODIUM HEXAFLUORIDE

C. L. Chernick, H. H. Claassen and Bernard Weinstock
J. Am. Chem. Soc. <u>83</u> 3165-3166 (July 20, 1961)   Letter

Abstract in ANL-6394, p. 34

*Illinois Institute of Technology

THE PREPARATION AND PROPERTIES OF PLATINUM HEXAFLUORIDE

Bernard Weinstock, J. G. Malm and E. E. Weaver
J. Am. Chem. Soc., in press

Platinum hexafluoride is the first simple hexavalent platinum compound that has been prepared. This was achieved by burning a platinum wire in a fluorine atmosphere adjacent to a cold surface with yields of up to 70%. The formula has been established by chemical analyses and vapor density. A solid transition occurs at $3.0°C$, 32.5 mm Hg pressure and the fusion point is $61.3°C$, 586 mm Hg. The heat of transition is 2140 cal mole^{-1} and the heat of fusion is 1080 cal mole^{-1}. The vapor pressure equations are: orthorhombic solid, log P(mm Hg) $= -3147.6/T-6.09$ log $T + 27.7758$; cubic solid, log P(mm Hg) $= 2528.7/T-4.080$ log $T + 20.6280$; liquid, log P(mm Hg) $= -5685.8/T-27.485$ log $T + 89.14962$. The boiling point is $69.14°C$, which is the highest boiling point of the known hexafluorides. The heat of vaporization at the boiling point is 7060 cal mole^{-1}. The liquid density is 3.826 g cc^{-1} at $64.3°C$. The density of the low temperature solid varies between $6.0-5.0$ g cc^{-1} in the range 77 to $254°K$. The vapor and liquid are red in color and the solids appear black. The molecule does not rotate freely in any of the condensed phases. Platinum hexafluoride is the least stable and most reactive of the known hexafluorides. The use of PtF_6 as a fluorinating agent at room temperature to form BrF_5, NpF_6 and PuF_6 is described.

URANIUM MONOSULFIDE. I. VAPORIZATION; THERMODYNAMICS AND PHASE BEHAVIOR

E. D. Cater, P. W. Gilles and R. J. Thorn
J. Chem. Phys. 35 608-618 (August 1961)

Abstract in ANL-6394, p. 35

URANIUM MONOSULFIDE. II. MASS SPECTROMETRIC STUDY OF ITS VAPORIZATION

E. D. Cater, E. G. Rauh and R. J. Thorn
J. Chem. Phys. 35 619-624 (August 1961)

Abstract in ANL-6394, p. 36

SPECIFIC INTERACTION BETWEEN Np(V) AND U(VI) IN AQUEOUS PERCHLORIC ACID MEDIA

J. C. Sullivan, J. C. Hindman and A. J. Zielen
J. Am. Chem. Soc. 83 3373-3378 (August 20, 1961)

Abstract in ANL-6394, p. 36

SOME OBSERVATIONS OF Np(VI) IN CHLORIDE SOLUTIONS
Donald Cohen and Brian Taylor
J. Inorg. and Nuclear Chem., in press   Note

Reports on the early work on the solution chemistry of neptunium contain reference to the instability of aqueous solutions of Np(VI) in the presence of chloride ion due to the reaction $2NpO_2^{++} + 2Cl^- \rightarrow 2NpO_2^+ + Cl_2$. In fact no satisfactory absorption spectrum of Np(VI) in HCl could be obtained in the near uv region. This study has shown that the reduction of Np(VI) in chloride media is very slow, not differing greatly from that expected from autoreduction and that a satisfactory absorption spectrum can be obtained. The accelerated reduction observed in the presence of a platinum electrode is shown to be caused by platinum itself being the reducing agent.

Np(VI) IN MOLTEN NITRATES
Donald Cohen
J. Am. Chem. Soc. 83 4094 (October 5, 1961)   Letter

There has been no direct evidence for the existence of Np(VI) in any molten salt system. However, a study of the solvent extraction of neptunium from the $LiNO_3$ - KNO_3 eutectic showed a large increase in the distribution coefficient upon the addition of NH_4NO_3. This fact could be explained by either a "salting effect" or by a valence change. This work has shown that Np(V) is oxidized to Np(VI) by the action of NH_4NO_3 in the $LiNO_3$-KNO_3 eutectic. The Np(VI) ion yields a brownish yellow color in molten nitrate with the important absorption peak at 11,230 A. The Np(VI) ion is not stable in the nitrate melt and in a matter of days will revert to the (V) state. The Np(VI) ion is also thermally unstable; at about 220° it is reduced to the (V) state, and at about 380° the Np(V) ion precipitates from the melt, probably as NpO_2. Ozone will also oxidize Np(V) to Np(VI) in nitrate melts.

RAMAN SPECTRA OF SOLIDS
J. R. Ferraro, J. S. Ziomek and G. A. Mack
Spectrochim. Acta 17 802-814 (October 1961)

Abstract in ANL-6394, p. 40

INTRODUCTION TO INORGANIC INFRARED SPECTROSCOPY
J. R. Ferraro
Proc. 5th Ann. Infrared Spectroscopy Inst., Canisius College, Buffalo, April 14-18, 1961, in press

Abstract in ANL-6349, p. 48

THE BASIS OF SELECTIVITY IN CHROMATOGRAPHY, ELECTRO-CHROMATOGRAPHY AND CONTINUOUS ELECTROCHROMATOGRAPHY
H. H. Strain
Anal. Chem., in press

Selective separations by chromatography, electrochromatography, and continuous electrochromatography depend upon the substances being separated and upon the migration conditions. The number of these differential migration conditions is enormous and indeterminate. There is no common basis for comparison of the selective separations, and there is no widely applicable basis for the prediction of the selectivity of particular systems. There is no systematic relationship between the separability and the molecular structure of various substances.

CONTINUOUS SEPARATION OF GASEOUS MIXTURES BY THERMAL GRAVITATIONAL DIFFUSION
H. D. Frame, Jr., J. R. Kuszewski, J. F. Binder and H. H. Strain
Anal. Chem., in press

For the continuous separation of gases by thermal gravitational diffusion, a rectangular diffusion cell (internal dimensions: 51 x 51 x 1 cm) has been constructed and tested. For the thermal gradient, one large wall was provided with a water jacket and cooled with tap water. The opposite large wall was heated against a specially wound, vertical electric furnace. Gas flow through the vertical diffusion chamber was horizontal. This arrangement provided three mutually transverse driving forces (thermal gradient, gravity, and gas flow).

Separations were made in diffusion cells with a structure-free diffusion chamber, with a chamber containing loosely packed silica fibers (vertically aligned), and with a chamber containing porous stainless steel barriers (also vertically aligned). The structure-free cell produced the best helium-nitrogen separation, the cell with silica fibers the best nitrogen-argon separation, and the barrier cell the best neon isotope enrichment.

No significant dependence of the separations upon flow was found in any of the cells with flow rates from no flow to about 9% of the total internal volume per minute (150 ml/min at room temperature).

SEPARATION OF RARE EARTHS BY LIQUID-LIQUID EXTRACTION
D. F. Peppard
The Rare Earths, ed. F. H. Spedding and A. H. Daane. New York:
John Wiley & Sons, Inc., 1961. pp. 38-54

The presentation is the expansion of material presented orally to
a group primarily interested in separations. It makes no pretense to
completeness in theoretical aspects and little pretense to completeness
in applications, except where they appear to be unusually promising.
For these reasons, considerable liberty has been taken in attempting
to predict the course of future developments in the liquid-liquid extrac-
tion fractionation of the rare earths.

THE RADIOCHEMISTRY OF NIOBIUM AND TANTALUM
E. P. Steinberg
Natl. Acad. Sci., Natl. Research Counc. Nuclear Sci. Ser.,
NAS-NS 3039 (August 1961)

This monograph lists general reviews of the inorganic and analytical
chemistry and radiochemistry of niobium and tantalum. It reviews in more
detail those features of niobium and tantalum chemistry of particular inte-
rest to radiochemists: the metallic state, soluble and insoluble salts,
behavior of tracer quantities, complex ions, solvent extraction behavior,
and ion exchange, as well as problems of sample dissolution and inter-
change with carrier and counting techniques. Detailed radiochemical
separation procedures of Nb and Ta from each other and from other elements
are given.

Miscellaneous

DEUTERIO-CAROTENOID PIGMENTS FROM FULLY DEUTERATED
GREEN ALGAE
H. H. Strain, Mary Thomas, H. L. Crespi and J. J. Katz
Biochim. Biophys. Acta 53 517-526 (1961)

Fully deuteriated crystalline α- and β-carotene and lutein have been
isolated from fully deuteriated algae, and solutions of deuterio-violaxanthin
and deuterio-neoxanthin have been obtained. Significant differences occur
in the visible absorption spectra of the deuterio-carotenoids as compared
to those of the hydrogen compounds, and these differences are discussed.
Ordinary and deuterio-β-carotene can be separated quite completely by
chromatography in activated magnesia, thus further illustrating the extent
to which certain properties are modified by extensive isotopic replacement.

SPECTRA OF ESCHSCHOLTZXANTHIN AND OTHER CAROTENOID PIGMENTS

H. H. Strain, M. R. Thomas and J. J. Katz

J. Organ. Chem., in press

Spectral absorption properties in the visible and in the infrared regions indicate that eschscholtzxanthin, with 12 conjugated double bonds, is more closely related to symmetrical dehydro-β-carotene, with a centrally-located single bond than to γ-carotene or to dehydro-carotenoids, with a double bond at the center of the molecule. Most of the absorption maxima of the common carotenoids in the infrared have been related to absorption by particular structural units.

EVIDENCE FOR EXCHANGEABLE HYDROGEN IN CHLOROPHYLL

J. J. Katz, M. R. Thomas, H. L. Crespi and H. H. Strain

J. Am. Chem. Soc., in press

The hydrogen in position 10 in chlorophyll is in a favorable position for enolization and should, therefore, be both labile and exchangeable. However, attempts by earlier workers to demonstrate such an exchange have been unsuccessful. Holt and Jacobs (1955), have in spite of this, interpreted the infrared spectra of chlorophylls a and b to indicate that chlorophyll a exists as the enol (the hydrogen at carbon atom 10 being enolized), whereas chlorophyll b exists in the keto form. If this interpretation is correct it should be possible to detect labile hydrogen in a but not in b.

Since chlorophyll a contains 72 hydrogen atoms, it did not appear feasible to detect the replacement of one hydrogen atom by deuterium by a direct examination of the infrared spectrum of the exchanged chlorophylls. Consequently, an indirect procedure, also depending on infrared spectrophotometry, was employed. Chlorophyll was treated with a large excess of CH_3OD; if exchange occurred, then deuterium was introduced into the chlorophyll. If this chlorophyll were then allowed to exchange with an equivalent amount of ordinary methanol, CH_3OH, exchange would result in a mixture of CH_3OH and CH_3OD. The ratio of CH_3OH to CH_3OD is easily determined in the infrared and the extent of exchange thus readily deduced. Exchange with chlorophyll in a solvent of low dielectric constant is slow but unmistakable. Both chlorophyll a and b were found quite conclusively to possess at least one exchangeable hydrogen atom and both substances were observed to exist predominately in the keto-form.

SOLID STATE SCIENCE DIVISION

THE SPECIFIC HEAT AT CONSTANT VOLUME, THE ENTROPY, THE INTERNAL ENERGY AND THE FREE ENERGY OF LIQUID He^4 BETWEEN 1.2 AND 2.9°K

O. V. Lounasmaa
  Cryogenics $\underline{1}$(4) 212-221 (June 1961)

Abstract in ANL-6349, p. 54

HOLE PHOTOCONDUCTIVITY IN KBr CRYSTALS CONTAINING Br$_2^-$ MOLECULE IONS

C. J. Delbecq
  Bull. Am. Phys. Soc. $\underline{6}$ 483 (November 1961)   Abstract
  J. Phys. and Chem. Solids, in press

After x-ray irradiation at liquid nitrogen temperature, holes are trapped by forming Br$_2^-$ molecule ions in KBr containing one of the impurities Ag$^+$, Tl$^+$, Ag$^+$, or NO$_2^-$. It has been found that the impurities act as efficient electron traps and increase the rate of production of Br$_2^-$. An investigation of the optical and paramagnetic resonance spectra of these crystals leads to an identification of the optical absorption bands of the Br$_2^-$ molecule ion. The Br$_2^-$ molecule ions are oriented along $\langle 110 \rangle$ directions in the crystal and have two principal absorption bands at 385 mμ and 750 mμ, both of which show anisotropic properties. Irradiation into either one of these bands with [0$\bar{1}$1] polarized light causes a redistribution of the ions among the six $\langle 110 \rangle$ directions in the crystal so that the population in [011] is considerably enhanced. Because of this change in population, the absorption as measured with [0$\bar{1}$1] light will decrease, and the absorption as measured with [011] light will increase. Photoconductivity has been observed upon bleaching with light of 750 mμ wavelength. The bleaching light used was polarized along either [0$\bar{1}$1] or [011]. Since continued bleaching at 750 mμ with [0$\bar{1}$1] light causes a decrease in the absorption to [0$\bar{1}$1] light, the observed photoconductivity decreases. If the plane of polarization of the bleaching light is rotated to [011], the photoconductivity rises and then decays to an equilibrium value. These changes in photoconductivity have been correlated with changes produced in the optical absorption spectra of Br$_2^-$.

PARAMAGNETIC NITROGEN OXIDES IN IRRADIATED POTASSIUM
HALIDES
Claude Jaccard
Phys. Rev. 124 60-66 (October 1, 1961)

Abstract in ANL-6394, p. 53

BORN-MODEL TREATMENT OF THE POLYMORPHIC TRANSITIONS
OF THE ALKALI HALIDES
M. P. Tosi and F. G. Fumi
J. Phys. and Chem. Solids, in press

The Born model of ionic solids is shown to account for the observed values of the work involved in the pressure transition of the potassium and rubidium halides from the NaCl to the CsCl structure, and for the heat absorbed in the thermal transition of cesium chloride from the CsCl to the NaCl structure, when one allows the Born repulsive parameters to depend on the crystal structure. These parameters are determined separately for the NaCl phase and for the CsCl phase of each compound from the observed values of the lattice parameter and of the compressibility of the two phases at the transition point. The motivation and physical significance of this procedure are discussed.

ON THE MIE-GRÜNEISEN AND HILDEBRAND APPROXIMATIONS TO THE EQUATION OF STATE OF CUBIC SOLIDS

F. G. Fumi and M. P. Tosi

An analysis is made of the conditions under which the equation of state of a cubic solid under hydrostatic pressure and its first volume derivative at constant temperature take the form given by either the Mie-Grüneisen or the Hildebrand approximation. The condition of validity of the vibrational or thermal formulation of the Mie-Grüneisen approximation is reduced to the existence of a purely volume-dependent characteristic temperature for the vibrational or thermal free energy of the solid. The analysis consists, then, of a search for temperature ranges where these restrictions on the functional form of the free energy, and the restrictions on the form of the internal energy imposed by the Hildebrand approximation, are satisfied for a non-metal in the quasi-harmonic approximation. The main results are as follows: (i) At temperatures somewhat above the Debye characteristic temperature for the (quasi-harmonic) high-temperature heat capacity at constant volume, it is appropriate to take as equation of state the vibrational Hildebrand equation; (ii) At somewhat lower temperature, this Hildebrand equation is generally more inaccurate than the corresponding Mie-Grüneisen equation; (iii) In the low-temperature T^3 region of the heat capacity, the equation of state reduces to the thermal Mie-Grüneisen equation. Some corollary results are obtained, within the quasi-harmonic approximation, on the temperature variation at constant volume of the Grüneisen parameters relating the explicit volume and temperature dependence of the vibrational and thermal free energy and of the entropy of a cubic solid, and (in an appendix) on the temperature variation of the Debye temperatures appropriate to the various thermodynamic functions of any non-metal. The available experimental and theoretical evidence on the anharmonic contributions to the thermodynamic functions of solids is briefly discussed, and points to the conclusion that their weight is quite small in the region of temperatures of interest for our analysis.

THEORY OF SLOW NEUTRON SCATTERING BY LIQUIDS. I.

Aneesur Rahman, K. S. Singwi and A. G. Sjölander
Phys. Rev., in press

Based on Van Hove's formalism, a general discussion of scattering in liquids has been given. The scattering cross section has been expressed in terms of velocity correlation functions; in particular, for the incoherent scattering cross section it is shown that in the Gaussian approximation for Van Hove's $G_s(\underline{r},t)$-function, only a knowledge of the velocity auto-correlation function $<\underline{v}(0) \cdot \underline{v}(t)>_T$ is necessary. The departure from the Gaussian approximation is expressed in terms of higher order velocity correlation functions. A derivation of an approximate formula for the width function of the Gaussian $G_s(\underline{r},t)$, suggested earlier by the authors, has been given. The frequency spectrum of the velocity auto-correlation function has been introduced, and it has been shown that, as a consequence of the fluctuation-dissipation relations, the spectral representation of the width function is formally identical with that obtained earlier for a harmonic solid. The first few moments of the energy transfer have been discussed. Some of these moments have been shown to satisfy certain relations which involve only experimentally observable quantities; and hence, these relations can be used as a check on the internal consistency of the experimental data.

STOCHASTIC MODEL OF A LIQUID AND COLD NEUTRON SCATTERING. II.

Aneesur Rahman, K. S. Singwi and A. G. Sjölander
Phys. Rev., in press

A simple model for the atomic motions in a liquid has been constructed based on the assumption that rapidly varying motions behave similarly to those in a solid, whereas slowly varying motions behave according to Langevin's equation for diffusion. This has been accomplished by writing the displacement of an atom as a sum of statistically independent "modes." Each "mode" is assumed to obey Langevin's equation for a harmonic oscillator with a certain frequency and a certain damping. To account for diffusion it is assumed that for "modes" below a certain frequency the harmonic restoring force is absent and thus these "modes" obey Langevin's equation for diffusion.

Based on this model Van Hove's $G_s(\underline{r},t)$ function is Gaussian and its width function has been calculated. The computed scattering cross section for neutrons has been compared with experimental data for water and for liquid lead.

PARAMAGNETIC RESONANCE OF SOME COPPER COMPLEX COMPOUNDS

J. A. McMillan and Bernard Smaller

THERMODYNAMICS AND STATISTICAL MECHANICS OF A THREE-LEVEL MASER

W. A. Barker

APPROXIMATE SOLUTION OF FICK'S DIFFUSION EQUATION

Tung Tsang

A THEORETICAL STUDY OF SIMPLE MANY-ELECTRON SYSTEMS
L. M. Sachs
ANL-6310 (May 1961)

A study has been made of the one-electron approximation for a number of simple many-electron systems. A review is given of the mathematical structure underlying the Hartree-Fock (HF), Unrestricted Hartree-Fock (UHF), and Projected Unrestricted Hartree-Fock (PUHF) approximations which may be utilized to obtain approximate solutions of the many-electron Schrödinger equation. The salient features of each method and their applicability to simple systems are outlined.

Solutions using the above approximations were obtained by the "self-consistent field" (SCF) method utilizing a finite set of Slater-type functions as an expansion set. This becomes a matrix procedure and involves the calculations of the matrix elements of the operators and the solution of the matrix (operator) equations. To this end, a number of computer programs were written for the IBM-704 digital computer and utilized to obtain the desired solutions.

Hartree-Fock solutions have been obtained for the atomic configurations $1s^2$, $1s^2 2s$, and $1s^2 2s^2$ for the range of atomic number $2 \leq Z \leq 10$. The UHF solution of lithium has also been obtained and the PUHF solution projected from it. A tabulation of the orbitals and two measures of their quality is given for all SCF solutions. Utilizing the HF, UHF, and PUHF solutions, the hyperfine splitting of the lithium ground state is calculated by first-order perturbation and found to be given best by the UHF method, approaching the experimental value. The diamagnetic susceptibilities of He, Li^+, Li^-, and Be have been calculated.

Hartree-Fock solutions have also been obtained for the ground states of F, F^-, Ne, Na^+, and Na, and for Na^+ and F^- in the presence of superposed charged spheres. A tabulation of the orbitals and their goodness is given for this series. The diamagnetic susceptibilities of the ten-electron systems has been calculated. The doublet separation of the fluorine ground states has been computed, and the hyperfine splitting of sodium and fluorine have been obtained.

A number of recommendations are offered regarding the SCF computational procedure.

CHEMICAL ENGINEERING DIVISION

KINETICS OF THE REACTION OF SULFUR TETRAFLUORIDE WITH URANIUM TRIOXIDE AND URANYL FLUORIDE

C. E. Johnson and Jack Fischer
J. Phys. Chem. 65 1849-1852 (October 1961)

The reaction between sulfur tetrafluoride and uranium trioxide, sulfur tetrafluoride and uranyl fluoride has been studied between 255° and 370° by following the change in weight of the solid phase using a thermo-balance. The rate of production of uranium hexafluoride is in agreement with the kinetics expected for reaction between a gas and a solid at a continuously diminishing spherical interface. The rate of reaction is temperature dependent. The activation energy for reaction of sulfur tetrafluoride with uranium trioxide and uranyl fluoride is respectively 6.0 kcal/mole and 32.0 kcal/mole. The rate of reaction with uranyl fluoride is dependent upon the partial pressure of sulfur tetrafluoride. A 1.55 exponent was found. A minimum gas velocity was found above which the reaction rate is independent of the velocity of gaseous reactant past the solid.

THE KINETICS AND MECHANISM OF THE THERMAL DECOMPOSITION OF PLUTONIUM HEXAFLUORIDE

Jack Fischer, L. E. Trevorrow and W. A. Shinn
J. Phys. Chem. 65 1843-1846 (October 1961)

The kinetics and mechanism of the decomposition of plutonium hexafluoride has been investigated and the rate of decomposition has been formulated as a concurrent first and zero order reaction with respect to plutonium hexafluoride pressure. It has been inferred that the decomposition, in the temperature range studied, proceeds by both a homogeneous and heterogeneous unimolecular decomposition. The heterogeneous decomposition occurs on the surface of plutonium tetrafluoride.

SOLUBILITIES IN LIQUID ZINC: ZIRCONIUM, NIOBIUM, MOLYBDENUM, PALLADIUM AND THORIUM

A. E. Martin, J. B. Knighton and H. M. Feder
J. Chem. and Eng. Data 6 (4) 596-599 (October 1961)

Abstract in ANL-6394, p. 56

STUDIES OF METAL-WATER REACTIONS AT HIGH TEMPERATURES.
I. THE CONDENSER DISCHARGE EXPERIMENT: PRELIMINARY
RESULTS WITH ZIRCONIUM

L. Baker, Jr., R. L. Warchal, R. C. Vogel and M. Kilpatrick*
ANL-6257 (May 1961)

The condenser-discharge method of conducting molten metal-water
reactions at high temperatures was refined. Two methods to measure energy
input to specimen wires and, therefore, to compute initial metal tempera-
tures were developed. Calculated metal temperatures were estimated to be
accurate to within 100 C. Two reaction cells were designed, one for opera-
tion at atmospheric pressure with water at room temperature, and the other
for operation at high pressure and with water at elevated temperature.
Means were developed to determine the surface area of metal exposed to
reaction and to determine the total extent of reaction. Pressure transducers
were used to record the rate of reactions.

The zirconium-water reaction was studied with initial metal tem-
peratures from 1100 to 4000 C with 30- and 60-mil wires in room-temperature
water. Initial pressures in these runs were the vapor pressures of water
at room temperature (20-30 mm). Runs were made with 60-mil wires in
water heated to 200 C (225 psi). Results in room-temperature water in-
dicated that the reaction became explosive at an initial metal temperature
of 2600 C. Below this temperature, 20% or less reaction occurred. At
higher water temperatures, reaction ranged from 40 to 70%. Runs in heated
water showed markedly greater reaction, reaching 50% for fully melted
metal at the melting point (1840 C). Results suggested that the rates of
both solid-state processes and the diffusion of water vapor through the hy-
drogen blanket surrounding reacting particles must be considered.

FLUORINE BOMB CALORIMETRY. I. THE HEAT OF FORMATION OF
ZIRCONIUM TETRAFLUORIDE

Elliott Greenberg, J. L. Settle, H. M. Feder and W. N. Hubbard
J. Phys. Chem. 65 1168-1172 (July 1961)

Abstract in ANL-6349, p. 59

FLUORINE BOMB CALORIMETRY. II. THE HEAT OF FORMATION OF
MOLYBDENUM HEXAFLUORIDE

J. L. Settle, H. M. Feder and W. N. Hubbard
J. Phys. Chem. 65 1337-1340 (August 1961)

Abstract in ANL-6394, p. 55

*Chemistry Division.

PURIFICATION OF FLUORINE BY DISTILLATION
Lawrence Stein,* Edgars Rudzitis and J. L. Settle
ANL-6364 (June 1961)

A vacuum-jacketed metal still for purifying half-pound quantities
of commercial fluorine is described. Operating characteristics of a proto-
type still and of the present still are reported. The distillate is analyzed
by an improved mercury-titration method, and a middle fraction of 99.9 per
cent or higher purity is collected. The impurities found in several cylinders
of commercial fluorine by infrared and mass spectrographic analyses in-
clude nitrogen, oxygen, carbon dioxide, nitrogen trifluoride, hydrogen fluoride,
carbon tetrafluoride, perfluoroethane, perfluoropropane, oxygen difluoride
and argon.

THE CADMIUM-URANIUM PHASE DIAGRAM
A. E. Martin, Irving Johnson and H. M. Feder
Trans. Met Soc. A.I.M.E. $\underline{221}$ 789-791 (August 1961)

Abstract in ANL-6349, p. 59

A RECORDING EFFUSION BALANCE FOR PHASE DIAGRAM INVESTI-
GATIONS: U-Cd, U-Zn, AND Ce-Zn SYSTEMS
Ewald Veleckis, C. L. Rosen and H. M. Feder
J. Phys. Chem., in press

An apparatus for the determination of the volatility of one com-
ponent of a system over a wide range of compositions is described. The
apparatus, which couples a Knudsen effusion crucible with a recording
balance, permits the investigator to make a rapid survey of phase boundary
locations and, simultaneously, to observe the decomposition pressures of
the phases involved. Three test systems were studied. The results cor-
roborated, modified or extended previously known phase diagrams. In the
U-Cd system only the one (known) intermetallic compound, UCd_{11}, was
found. Between 420°C and 500°C the U-Zn system study disclosed a single
intermetallic phase, $UZn_{8.5-\sim12}$. Only the end member of this solid solu-
tion range, $UZn_{8.5}$, had been previously described. In the Ce-Zn system
eight intermetallic phases were found: $CeZn_{11}$, Ce_2Zn_{17}, $CeZn_{\sim7}$, $CeZn_{3.8-6.2}$,
$CeZn_2$, $CeZn$, Ce_2Zn and Ce_4Zn. Of these eight phases, three had been known,
two had been suspected, and three had been indicated by indirect evidence.

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*Chemistry Division.

PREPARATION OF HIGH-DENSITY, SPHERICAL THORIUM OXIDE PARTICLES WITH UP TO 10 ATOM PER CENT URANIUM

C. E. Crouthamel, W. G. Knapp, S. B. Skladzien and J. W. Loeding
ANL-6340 (June 1961)

A relatively simple method has been evolved in which up to about 10 atom per cent uranium may be homogeneously distributed throughout a spherical particle of thorium dioxide. The particle size can be varied from about one micron to fifty microns in diameter, and the final particle density is very close to the theoretical maximum of 10 g/cm^3. This material described here was designed for use as a fuel in a boiling water slurry reactor. Some of the important characteristics of the individual particles for this application were the shape of the particles, the size, the density, and the homogeneity of distribution of fissile atoms in the thoria matrix. The fabrication process appears to be adaptable to large-scale production. About 100 lbs of fired spherical material have been produced by this method.

PROCESS VESSEL DESIGN FOR FROZEN-WALL CONTAINMENT OF FUSED SALT

R. W. Kessie, J. D. Gabor, W. J. Mecham and A. A. Jonke
ANL-6377 (August 1961)

The feasibility of the containment of a molten sodium fluoride-zirconium fluoride salt at 550 C in vessels with a protective inner liner of frozen salt of the same composition was demonstrated. Two different designs were used for supplying internal heat directly to the molten salt: induction heating and electrolytic heating. The test equipment was of a size practical for processing zirconium-uranium alloy reactor fuel by the Fused Salt Process.

METALLURGY DIVISION

A NEUTRON DIFFRACTION STUDY OF KRYPTON IN THE LIQUID STATE
G. T. Clayton and LeRoy Heaton
ANL-6112 (June 1961)

A study was made of the neutron diffraction patterns obtained from krypton in the liquid state under seventeen conditions of temperature and pressure between 117°K and 210°K. The diffraction patterns were obtained by use of one of the neutron diffractometers of the Metallurgy Division of the Argonne National Laboratory.

Neutrons of 1.05-A wavelength were selected from a beam of neutrons from the CP-5 reactor by reflection from the (111) planes of a single copper crystal, and this nearly monochromatic beam was incident upon the sample. The patterns obtained at 117°K, 133°K, 153°K, 183°K, and 210°K were repeated at least three times to obtain more reliable patterns for making further analysis. These diffraction patterns are the first ever reported for liquid krypton, and they show four well-defined prominences at the lower temperatures and three at the higher temperatures.

This program required the development of apparatus with unique characteristics which would permit the containment of the sample at elevated pressures. The krypton under study filled a volume of 0.660-in. diameter and 1.5-in. height in a cell fabricated from an aluminum single crystal grown especially for this research. This cell withstood pressures up to 55 atmospheres while contributing to the cell scattering of neutrons in an amount lower than ever reported by anyone for this kind of work. The cell was housed in an evacuated cryostat and kept at temperatures ranging from 117°K to 210°K at values constant to within 0.05°K by electronic controlling circuits, and the original charge was contained in a closed system for the duration of the experiment. After appropriately correcting the diffraction patterns for effects of background scattering, incoherent scattering, and empty cell scattering, the intensity data were Fourier transformed to give the atomic radial distribution function at each of the temperatures. The calculations were performed by use of the computer GEORGE of the Argonne National Laboratory. The speed of this computer made it feasible to investigate the sensitivity of the resultant distribution functions to arbitrary adjustment in the input data. Studies were made of the effect on the distribution of (1) terminating the calculation at arbitrary points, (2) using different values of incoherent scattering correction, (3) using different estimates of independent scattering, and (4) modifying the data by arbitrary exponential factors.

Atomic distributions at all temperatures showed nondiscrete first peaks, indicating that no molecule is formed. For the five temperatures at which the liquid was carefully studied, the positions of the first peaks and the number of nearest neighbors to which the first peak areas correspond are as follows: 117°K, 4.02 A, 8.4 atoms; 133°K, 4.03 A, 7.9 atoms; 153°K, 4.08 A, 7.0 atoms; 183°K, 4.10 A, 6.4 atoms; 210°K, 4.20 A, 4 atoms. The number of nearest neighbors was found to be closely proportional to the density of the liquid.

By use of the series of distribution functions calculated from arbitrarily terminated data, it was shown that the width of the potential bowl of liquid krypton is in approximate quantitative agreement with that of the Lennard-Jones potential. This series of patterns also indicate that some relevant information remains unobserved beyond the range of experimental observation.

THE ZIRCONIUM-RICH CORNERS OF THE TERNARY SYSTEMS
Zr-Co-O AND Zr-Ni-O
    M. V. Nevitt and J. W. Downey
        Trans. A.I.M.E. 221 1014-1017 (October 1961)

    Abstract in ANL-6245, p. 63

FACTORS CONTROLLING THE OCCURRENCE OF LAVES PHASES AND
AB_5 COMPOUNDS AMONG TRANSITION ELEMENTS
    A. E. Dwight
        Trans. A.S.M. 53 479-500 (1961)

Intermetallic compounds were the subject of this investigation, and the following examples were studied: 32 $MgCu_2$-type, 10 $MgZn_2$-type, one $MgNi_2$-type, 27 $CaCu_5$-type, and one UNi_5-type. The interrelation of crystal structures was shown for the five families. The effect of atomic size was studied. It was found that Laves phases may form in the range of Goldschmidt radius ratios from 1.05 to 1.68 and that the radius ratio has little influence on the structure type. The positions of the partner elements in the periodic table are the dominant factor which controls the formation of any one of the three Laves phases, but the occurrence of the Laves phases as a group is dependent upon the ability of the partner atoms to undergo the necessary contraction or expansion required to approach the effective radius ratio 1.23.

The $CaCu_5$-type compounds exist in the radius ratio range from 1.29 to 1.61. Below this range UNi_5-type compounds are formed.

ALLOYING BEHAVIOR OF COLUMBIUM
   A. E. Dwight
      Columbium Metallurgy, ed. D. L. Douglass and F. W. Kunz. New
      York: Interscience Publ., Inc., 1961. pp. 383-404

   Abstract in ANL-6298, p. 67

STRUCTURES AND PROPERTIES OF URANIUM-FISSIUM ALLOYS
   S. T. Zegler and M. V. Nevitt
      ANL-6116 (July 1961)

   A study was made of the phase relations and the properties of
uranium-fissium alloys which have compositions bracketing that intended
for the first core loading of Experimental Breeder Reactor II. The fis-
sium aggregate in the alloys consisted of the elements Zr, Nb, Mo, Ru,
Rh, and Pd. Phase relations are shown to parallel closely those in the
dominant U-Mo-Ru ternary system. The uranium gamma phase is stabi-
lized down to 552°C, while the beta phase is entirely suppressed at high
fissium contents. Certain crystallographic data are given and the minor
phases that occur in the alloys are identified. In cast and gamma-quenched
alloys the retention of the high-temperature gamma phase produced low
hardness and low density. The thermal expansion behavior of the alloys
is shown to be dependent upon composition and prior thermal history.
Thermal conductivity data are presented for uranium and the uranium-
fissium alloys. The thermal conductivities of the alloys decrease with
increasing fissium concentration.

DIFFUSION IN URANIUM, ITS ALLOYS, AND COMPOUNDS
   S. J. Rothman
      ANL-5700, Part C (May 1961)

   This paper reviews laboratory diffusion studies. It attempts to
evaluate them in the light of present-day diffusion theory to decide which
set of data is to be preferred in the case of conflict and to indicate what
fields of study need further work. The diffusion work considered in this
review can be divided into three major classes according to the type of
sample used. These are: 1. Diffusion in a single-phase solid. The
quantity of interest here is the diffusion coefficient which is usually ob-
tained from the variation of the concentration of the diffusant with distance
in the sample. 2. Diffusion in a multiphase solid. The quantities of
interest here are the rate constants for the growth of the intermetallic
layers. 3. Diffusion of gas in a solid. The quantity of interest is again
the diffusion coefficient; in this case it is obtained from measuring the
amount of gas that diffuses out of the solid as a function of time.

A LOW-TEMPERATURE PHASE TRANSITION IN ALPHA URANIUM

E. S. Fisher and H. J. McSkimin*

Phys. Rev. 124 67-70 (October 1, 1961)

A phase transition in alpha uranium at $42°K \pm 1°K$ is indicated by measurements of the single-crystal elastic moduli using ultrasonic pulse techniques. The indicated transition coincides closely to anomalies noted in the thermal expansion, Hall coefficient, electrical resistivity, and thermoelectric power. An examination of the literature specific heat data shows that the transition is associated with an inflection point in the entropy vs temperature curve. On the basis of the elasticity and available lattice parameter data it is postulated that the temperature dependence of the y positional parameter becomes relatively large in this range and the phase instability is a result of a change in the nature of certain interatomic bonds.

LATTICE DYNAMICS OF ALPHA URANIUM

D. O. Van Ostenburg

Phys. Rev. 123 1157-1162 (August 15, 1961)

Abstract in ANL-6394, p. 61

ADIABATIC ELASTIC MODULI OF SINGLE CRYSTAL ALPHA ZIRCONIUM

E. S. Fisher and C. J. Renken

J. Nuclear Materials, in press

The elastic moduli of alpha zirconium at 20°C have been derived from measurement of the velocities of ultrasonic waves using the phase comparison technique of McSkimin. The values computed for a density of 6.505 g/cc are as follows in units of 10^{12} dynes/cm²: $c_{11} = 1.435 \pm 0.002$; $c_{12} = 0.725 \pm 0.002$; $c_{13} = 0.654 \pm 0.002$; $c_{33} = 1.649 \pm 0.002$; $c_{44} = 0.3217 \pm 0.0003$.

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*Bell Telephone Laboratories

CRYSTALLINE THORIUM BORATE
Y. Baskin,* Y. Harada* and J. H. Handwerk

Crystalline thorium borate (ThB_2O_5) has been synthesized by the reaction of thoria with boric oxide at elevated temperatures; crystals of size suitable for single-crystal X-ray studies were grown in molten boric oxide at 1300°C. Crystallographic, chemical, and some physical properties of thorium borate crystals are presented, as well as the results of a preliminary phase study of the system ThO_2-B_2O_3. Amorphous thorium borate gel, formed by reaction of aqueous solutions of borax and thorium nitrate, is transformed into crystalline thorium borate and thoria on firing at elevated temperatures. Boric oxide does not appear to react with either zirconium oxide or uranium dioxide to form any crystalline compounds. The fact that uranium dioxide does not react with boric oxide, but thoria does, affords a basis for effecting a chemical separation of the two oxides from thoria-urania solid solutions. Preliminary data on the reactions between boric oxide and different thoria-urania solid solutions are given.

CORROSION OF ZIRCONIUM ALLOYS IN SUPERHEATED STEAM
R. D. Misch and Sherman Greenberg

Presently available zirconium alloys do not have adequate corrosion resistance for nuclear superheater conditions, such as 540°C and 600 psi steam. Three avenues of approach have been followed in an effort to produce alloys of adequate corrosion resistance: 1. Compensating effects in the corrosion product. 2. Promotion of electronic conductance of corrosion product without accompanying deleterious effects. 3. Catalysis of the combination of corrosion product hydrogen at the oxide surface.

Although it is apparent that the actual situation is more complex than the above simplifications suggest, progress has in fact been made. Alloys have been produced which have corrosion rates of about 1 mil/year. Consideration must be given to the dependence of corrosion rate on heat treatment and to possible embrittlement of alloys by the absorption of corrosion product hydrogen.

*Armour Research Foundation

CORROSION STUDIES OF TERNARY ZIRCONIUM ALLOYS IN HIGH-TEMPERATURE WATER AND STEAM

R. D. Misch and C. Van Drunen
ANL-6370 (July 1961)

The alloying of zirconium to improve corrosion resistance has an empirical basis, and satisfactory explanations for the alloying effects are not available. A theory of compensating valencies in the corrosion oxide is proposed, in which cations of lower and higher valence than zirconium (+4) are present in ratios such that electrostatic neutrality is ensured. An example is an alloy containing equimolar amounts of scandium (+3) and niobium (+5).

A number of zirconium alloys were prepared in which scandium or yttrium were paired with elements capable of a +5 or +6 valence. The ternary alloys containing scandium were superior to the alloys containing yttrium. The alloys containing scandium plus molybdenum, tantalum, or tungsten had relatively long lifetimes in steam at 540°C and 600 psi as compared with other alloy combinations, including Zircaloy-2. A quenched alloy containing 0.025 w/o Sc and 0.053 a/o Mo (that is, 0.05 mol % of each additive) corroded approximately according to a cubic law up to 758 hr, at which point the rate suddenly increased in a manner suggesting hydrogen damage.

Examination of the oxide film from alloys containing scandium and molybdenum showed only monoclinic ZrO_2. It is believed that stabilization of this form of ZrO_2 instead of the cubic or tetragonal forms is a factor in promoting corrosion resistance. In this way the protective character of the film can be improved independently of the addition of cathodes.

THE POLARIZATION OF METALS IN DISTILLED WATER

F. E. DeBoer, J. E. Draley and C. A. Youngdahl
J. Electrochem. Soc. 108 167c (August 1961)   Abstract

A method of determining the polarization behavior of metals in distilled water has been developed. Difficulties are encountered due to potential gradients in the water, double layer capacity effects, and the influence of the polarizing current on the corroding interface. The procedures used to overcome these difficulties are described, and representative polarization curves are presented.

GLASSY MATERIALS FOR POTENTIAL NUCLEAR FUELS AND CONTROL
MATERIALS
J. H. Handwerk
Glass Bearing Fuels. Notes from the Meeting. U.S.A.E.C. Germantown, Md., January 31, 1961. TID-12617, p. 5

Of immediate interest in the application of glassy materials in nuclear energy are four primary areas of study: (1) the use of a fissile material bearing glass as a single phase fuel, (2) the use of a fissile bearing glass in admixture with crystalline fissile materials as a dispersion medium and sintering agent, (3) the use of a glass containing one or more high thermal cross section materials as a single phase control material, and (4) the use of a glass, in admixture with crystalline high cross section control materials as a dispersion medium or bonding agent.

Preliminary investigations were instigated to develop stable glasses which would contain a large amount of urania or which would contain a quantity of high cross section oxides such as Eu_2O_3, Gd_2O_3, or Dy_2O_3. Compositions in the various glass systems such as the alkali-alkaline earth-silica systems were smelted with varying amounts of urania and with up to 15 weight per cent of such oxides as Eu_2O_3, Gd_2O_3 and Dy_2O_3. These compositions were subjected to devitrification softening point, and volatility studies, and the initial results indicate that glasses containing 30 to 50 w/o can be formulated which will resist devitrification for periods of seven days over a temperature range of 300° to 900°C. Glasses containing over 50 w/o urania were found to contain undissolved urania following smelting or were found to crystallize when heated for prolonged periods of time over a temperature range of 300° to 900°C. Those compositions which contained up to 15 w/o of either Eu_2O_3, Gd_2O_3 or Dy_2O_3 were found to resist devitrification when heated for seven days over a temperature range of 300° to 900°C.

Glass compositions were also used to bond UO_2, UC and B_4C. Specimens were formed by either cold pressing followed by sintering, or by warm pressing at 400° to 600°C. These initial studies produced specimens of rather low densities, however, using modified procedures it may be possible to produce ware of high density at low temperatures. As high density is not a criteria for a neutron absorbing material, the low densities produced appeared to be suitable for a control material. These studies established that mixtures of B_4C and glass could be formed and sintered into rods having good mechanical strength, and by sintering a mixture of B_4C and glass in place in stainless steel tubes, a set of control rods was made for the ANL critical experiment.

Present studies are continuing to (1) establish compositional limits of the fuel bearing glasses, and to evaluate the viscosity gradient, temperature stability, and compatability of these glasses with cladding materials,

(2) study compaction and pressing characteristics of glass and fuel mixtures to achieve highest densities, (3) evaluate the use of glass as a solvent or as a bond for crystalline control materials, and (4) study the stability of promising compositions under irradiation.

HOOD VENTILATION IN ARGONNE'S PLUTONIUM FUEL FABRICATION FACILITY. A CRITICAL REVIEW

R. M. Mayfield and H. Bairiot

Abstract in ANL-6394, p. 76

A COMPARISON OF SEVERAL METHODS FOR THE PHOTOGRAPHIC DETECTION OF THERMAL NEUTRON IMAGES

Harold Berger

Several methods of photographic detection of thermal neutron images have been studied and compared with regard to photographic speed, relative neutron-gamma response and image sharpness qualities. All these comparisons have been made using a low gamma content, monochromatic neutron beam (1.05A) having an intensity of 3×10^5 $n/cm^2/sec$. The fastest speed, best neutron-gamma response of the direct exposure methods, and good image sharpness have been obtained using a direct exposure method employing a B-10 loaded scintillator and Type F x-ray film. Gadolinium and rhodium intensifier screens, with type KK film, have also yielded good results by the direct exposure method (film and intensifier screens exposed to the neutron beam together). Results are also reported for the use of indium, cadmium, silver and gold screens. The transfer method, in which the photographic film is exposed only to the radioactive decay radiation of an image-carrying metallic screen, and not to the neutron beam itself, has been found useful not only because no effect on the image is produced by gamma radiation in the neutron beam, but also because improved image sharpness has been obtained. In the cases of indium and gold (the two materials which have been found useful for the transfer method in the neutron flux available for this study), the improved image resolution obtained with the transfer method has been shown to result from the fact that the high energy prompt (n,γ) radiation is not detected on the film.

RADIATION RESISTANT, REMOTELY OPERATED, HIGH CAPACITY SPRING BALANCE
J. E. Ayer and G. J. Pokorny
Rev. Sci. Instr., in press

A balance with a weighing range between 9 and 24 kg has been tested. The use of radiation resistant sensing devices make it acceptable for use in hot cell applications. With an unrefined read-out method the system sensitivity, including resolution of chart reading by eye, at 65% confidence limits is ±10 g in 2.5-kg intervals. Over the entire 9- to 24-kg range the linearity of the system is such that the sensitivity is reduced to ±17 g. It is predicted that digital indication of weight and use of a low inertia, low friction indicating system will double the system sensitivity.
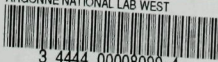
DEVELOPMENT OF REMOTE METALLOGRAPHIC TECHNIQUES FOR IRRADIATED MATERIALS
R. Carlander
ANL-6316 (September 1961)

A remote metallographic facility has been in operation at Argonne National Laboratory since 1954. During that period of time, many new techniques relative to better contamination control and equipment operation have been developed. Further improvements will continue to be made in the normal evolvement of the operational procedures. The techniques used for microscopic examination of irradiated materials have been standardized with variations only in the final polishing steps, and detailed procedures are given for several alloys. The procedures used for macroscopy vary from sample to sample, and new procedures are developed as required to suit each particular problem.